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ELASTIC ANOMALIES IN INTERMEDIATE VALENCE RARE-EARTH INTERMETALLICS

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The bulk moduli c_B of Rare earth (R) and Actinide (A) intermetallics (R-Cu₂Si₂, R-Rh₂, R-Ru₂, R-Be₁₃, A-Be₁₃) have been determined by means of Brillouin scattering. A softening of c_B is found in EuCu₂Si₂, YbCu₂Si₂, CeCu₂Si₂ and CeRh₂, but not in UBe₁₃. CeRu₂ shows a c_B twice as large as that of LaRu₂.

By means of an improved technique for Brillouin scattering, using a 3 + 3 pass tandem Fabry–Perot interferometer according to Sandercock [1], it has now become feasible to reliably measure bulk transverse and longitudinal acoustic sound velocities of metals. Thus we are able to determine the bulk modulus of both single crystals and polycrystals [2].

With the help of this method we have determined the bulk moduli c_B of various polycrystalline Rare earth (R) and Actinide (A) intermetallics (R-Cu₂Si₂, R-Ru₂, R-Rh₂, R- and A-Be₁₃) with particular emphasis on intermediate valence effects. These should show up most pronounced in the bulk modulus because of the volume changes associated with $4f^n \leftrightarrow 4f^{n-1} + e^-$ valence fluctuations. All measurements were carried out at room temperature. The c_B values of each series of compounds (except R-Ru₂) have been plotted as functions of Q/V , where Q is the valence of the R or A ion and V is the unit cell volume. For stable valent compounds this should yield a linear dependence of c_B on Q/V , whereas the intermediate valence effects are indicated by deviations from this behavior.

Fig. 1 shows the results for R-Cu₂Si₂. Contrary to the stable valent R-Cu₂Si₂ the compounds EuCu₂Si₂ and YbCu₂Si₂ exhibit a softening of the bulk modulus of 32 and 43%, respectively, with respect to the reference line. This softening is attributed to the $4f^n \leftrightarrow$

$4f^{n-1} + e^-$ configurational mixing near the Fermi level [3–5] which causes the R ions, and thus in turn the lattice, to become more compressible. A comparison of our c_B values with those determined by X-ray diffraction under hydrostatic pressure [6–8] as given elsewhere [2] shows good agreement within the error bars.

The 14% softening of c_B of CeCu₂Si₂ may also be associated with a valence instability effect, but even more so it may be indicative of the electron–phonon

Fig. 1. Bulk modulus c_B of R-Cu₂Si₂ vs. the ratio Q/V of the valence Q of the Rare earth (R) ion and the unit cell volume V of the lattice. The stable-valent R-Cu₂Si₂ follow a linear c_B vs. Q/V relation.

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coupling in this “heavy-fermion” superconductor ($T_c = 0.5$ K [9]). The large volume dependence of the spin-fluctuation or Kondo temperature ($T^* = 10$ K [9]) has recently been considered as the dominant electron–phonon coupling mechanism in CeCu_2Si_2 [10]. The softening of c_B is consistent with such a Kondo volume collapse-type of electron–phonon interaction.

An extraordinary situation is found in the case of the superconductor CeRu_2 ($T_c = 6.2$ K [11]). It turns out that c_B of CeRu_2 is twice as large as that of the reference compound LaRu_2 (CeRu_2 : $c_B = 134$ GPa, LaRu_2 : $c_B = 63$ GPa). An explanation for this behavior can be found in the valence of CeRu_2 , which was determined by L_{III} X-ray absorption as $v = 3 + \nu = 3.3$ [12]. On the basis of this valence and the superconducting properties of CeRu_2 the Ce ion may be considered as in the α' -Ce type phase [13], where the valence is not pressure-dependent ($d\nu/dp = 0$). Thus in eq. 1

$$\frac{1}{c_B} = - \frac{1}{V} \frac{\partial V}{\partial p} \bigg|_\nu - \frac{1}{V} \frac{\partial V}{\partial \nu} \bigg|_p \frac{d\nu}{dp}, \quad (1)$$

the second term vanishes and therefore CeRu_2 should show at least no softening in c_B , because the first term represents the “isovalent” contribution to the compressibility. Moreover, in the α' -phase of Ce the valence becomes reduced as compared to the α -phase of Ce [13], implying an increased volume of the Ce atom. Consequently, the α' -Ce type ion in CeRu_2 acquires a larger volume as compared to an α -Ce type ion. On the other hand, the unit cell volume of CeRu_2 is 6% smaller with respect to LaRu_2 (LaRu_2 : $V = 457 \text{ \AA}^3$, CeRu_2 : $V = 428 \text{ \AA}^3$ [14]). As a consequence of the “external” and the very large “internal” pressure the bulk modulus of CeRu_2 exceeds that of LaRu_2 by a factor of two.

CeRh_2 has a valence of 3.24 [12], which raises the

question whether it is α -Ce or α' -Ce like [13] and might exhibit a similar behavior as CeRu_2 . The bulk modulus of CeRh_2 has been determined together with that of La- , Y- and LuRh_2 . The c_B vs. Q/V plot in fig. 2 shows a 20% softening of c_B of CeRh_2 with respect to the reference line of the stable valent R-Rh_2 . Hence we conclude that the behavior of CeRh_2 resembles that of the α -Ce phase where the valence increases with increasing external pressure [13], so that the softening of c_B is caused by the second term in eq. 1.

The series of R- and A-Be_{13} intermetallics has been investigated in order to clarify whether c_B of intermediate valent CeBe_{13} is indeed larger than that of LuBe_{13} [15] and to determine c_B of the supposed p-wave superconductor UBe_{13} [16] ($T_c = 0.85$ K [17]). The results of c_B vs. Q/V are shown in fig. 3. The overall magnitude of the c_B values in fig. 3 is of the order of $c_B = 100$ GPa of Be. This is due to the fact that the Be atoms form cages around the R or A ions and in first order dominate the elastic behavior. For the presently available data a tentative reference line has been indicated in fig. 3 by the dashed line. The scaling between R- and A-Be_{13} compounds has to be taken with some caution as seen by the behavior of their ($q \approx 0$) optical phonon frequencies $\hbar\omega_{\text{opt}}$ vs. Q/V [18]. (While the stable valent R-Be_{13} show a linear variation of all $\hbar\omega_{\text{opt}}$ vs. Q/V , particularly ThBe_{13} shows non-systematic deviations from this behavior.) The non-softening of c_B of intermediate valent CeBe_{13} at 300 K is somewhat puzzling, especially in view of its anomalous temperature dependence upon cooling below 300 K [15]. While the overall softening of c_B of CeBe_{13} between 300 and 0.5 K has been attributed to the effect of approaching a second-order γ - α Ce type phase transition because of

Fig. 2. c_B vs. Q/V of R-Rh_2 (R = rare earth).

Fig. 3. c_B vs. Q/V of R- and A-Be_{13} (R = rare earth, A = actinide).

the Ce dilution in the Be matrix [15,19], a magnetoelastic coupling of the volume strain to two distinct electronic states (possibly reminiscent of Γ_7 – Γ_8 crystalline electric field excitations) has been used to explain the minimum in the temperature dependence of c_B near 135 K [20].

The “heavy-fermion” superconductor UBe_{13} shows no anomaly in c_B , even when assuming a horizontal reference line for the A – Be_{13} . This result is in contrast to the behavior of $CeCu_2Si_2$ and may be indicative of an electronic superconducting transition of UBe_{13} due to an interaction different from the usual electron–phonon interaction [16].

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